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NUCLEAR CHEMISTRY RESEARCH

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I. INTRODUCTION

In our 1971 Proposal for Continuation, the following plans for calendar 1972 were laid out (not necessarily in this order):

1. To complete analysis of the gamma-gamma angular correlation data on levels in ^{109}Cd . This work is still in progress. See Section A of the present progress report, below.
2. To complete the collection of pre-equilibrium charge state distribution (CSD) data for the recoils from the $^{10,11}\text{B}(d,d)$ reaction. This has been accomplished, as reported in Section B1, below.
3. An investigation of equilibrium CSD's in targets of different chemical structures to search for the existence of a medium effect. This work is in its final stages and is described in Section B2, below.
4. To determine the equilibrium CSD of Be recoils. This work is in progress using recoils from the $^{11}\text{B}(d,\alpha)$ reaction, as described in Section B3, below.
5. To determine the pre-equilibrium CSD of a nuclear reaction recoil at significantly higher velocity than those observed in the past. This work has also been begun on the $^{11}\text{B}(d,\alpha)$ reaction as described in Section B3, below.
6. To initiate the use of position-sensitive detectors in the CSD experiments. The carrying out of these plans is described in Section B3, below.

7. (This goal was implicit in last year's proposal.) To continue with calculations based upon a model for recoil CSD's. This work is described in Section C, below.
8. To test a novel method of tritium enrichment in water by electrolysis. This work is described in Section D, below.
9. To continue the development of computer programs pertinent to our research program. Accomplishments in this area are described in Section E, below.
10. To complete the construction of an evaporation system for target preparation in our laboratory. This has not been accomplished, as described in Section F, below.

II. SUMMARY OF RESEARCH

A. Gamma-Ray Spectroscopy

The gamma-ray coincidence and angular correlation experiments reported upon in last year's report have been completed and the data have been analyzed. Figure A1 shows the level scheme we have deduced for ^{109}Cd . Levels marked with an asterisk in the figure are new. From the directional correlation measurements, values of a_2 and a_4 coefficients were obtained by least-square fitting of the data. The results are summarized in Table AI and the proposed level scheme is shown in Figure A1. A paper on this work is in preparation.

Table AI - Summary of Gamma-Gamma Directional Correlation Results

Gamma Ray Energies (keV)	Experimental Coefficient		Assigned Sequence	Quadrupole Fraction
	a_2	a_4		
203.3 - 334	0.341 ± 0.018	0.531 ± 0.027	$\frac{5}{2} (M1) \frac{7}{2} (D, Q) \frac{5}{2}$	$0.4 < Q < 0.63$
203.3 - 216	1.044 ± 0.046	1.834 ± 0.073	$\frac{5}{2} (M1) \frac{7}{2} (D, Q) \frac{5}{2}$	_____
203.3 - 424.5	0.981 ± 0.085	1.241 ± 0.130	$\frac{5}{2} (M1) \frac{7}{2} (D, Q) \frac{9}{2}$ or $\frac{5}{2} (M1) \frac{7}{2} (D, Q) \frac{7}{2}$	_____
203.3 - 619.3	0.281 ± 0.088	-0.626 ± 0.118	$\frac{5}{2} (M1) \frac{7}{2} (D, Q) \frac{9}{2}$	$0.95 < Q < 1$
203.3 - 1148.5	0.179 ± 0.126	-0.602 ± 0.177	$\frac{5}{2} (M1) \frac{7}{2} (D, Q) \frac{9}{2}$	$0.0 < Q < 0.4$
288 - 177.0	-0.056 ± 0.046	2.664 ± 0.082	$\frac{5}{2} (M1) \frac{5}{2} (D, Q) \frac{7}{2}$	$0.96 < Q < 0.99$
288 - 650.1	0.337 ± 0.126	-0.591 ± 0.189	$\frac{5}{2} (M1) \frac{5}{2} (D, Q) \frac{5}{2}$	$0.67 < Q < 1$
288 - 374.8	0.64 ± 0.16	4.10 ± 0.30	$\frac{5}{2} (M1) \frac{5}{2} (D, Q) \frac{5}{2}$	_____

Figure A1

B. Charge States of Recoils

1. Recoils from $^{11}\text{B}(\text{d},\text{d})^{11}\text{B}$ Elastic Scattering

During the past year equilibrium and pre-equilibrium charge-state distributions (CSD's) have been measured for ^{11}B recoils at 11.1×10^8 cm/sec from $^{11}\text{B}(\text{d},\text{d})^{11}\text{B}$ elastic scattering in natural boron targets ranging in thickness from 0.03 to $54.20 \mu\text{g}/\text{cm}^2$. The experimental arrangement was the same as has been described previously (NYO-2771-40) except that beam divergence was limited by modifications to the beam-line slit system. Thickness determination of the natural boron targets is discussed in Section F.1 below.

As discussed last year, it was found that ^{11}B recoils in the 2.14 MeV excited state, produced at 6.98 MeV from the $^{10}\text{B}(\text{d},\text{p})^{11}\text{B}(2.14)$ reaction, interfered with the 7.00 MeV (11.1×10^8 cm/sec) ^{11}B recoils from $^{11}\text{B}(\text{d},\text{d})^{11}\text{B}$ elastic scattering. Since the energy resolution of the ^{11}B recoils at 7.00 MeV was 120-180 keV, the amount of interference of the $^{11}\text{B}(2.14)$ recoils to the recoil charge-state spectra had to be determined. This was done by using two targets of known thickness which had different isotopic ratios of ^{10}B to ^{11}B . From the recoil charge-state spectra, it was found that the ratio of the cross-section for the production of ^{11}B recoils from $^{11}\text{B}(\text{d},\text{d})^{11}\text{B}$ elastic scattering to the cross-section for the production of $^{11}\text{B}(2.14)$ recoils from the $^{10}\text{B}(\text{d},\text{p})^{11}\text{B}(2.14)$ reaction at $\theta_{\text{LAB}} = 15^\circ$ for 14.40 MeV incident deuterons was 34.48 ± 6.85 . Thus in a recoil spectrum from a natural boron target, only $0.71 \pm 0.14\%$ of the counts from the integrated peak at 7.00 MeV can be ascribed to the $^{10}\text{B}(\text{d},\text{p})^{11}\text{B}(2.14)$ interference. This value is well within the counting statistics for the charge-state data from $^{11}\text{B}(\text{d},\text{d})^{11}\text{B}$ elastic scattering and thus can be neglected.

It was found that $^{11}\text{B}^{2+}$ recoils were present to the extent of 0-2% of the total number of observed recoils. Since charge-state distributions with reasonable statistics (< 5%) from thin targets require large amounts of accelerator time, and since only three ^{11}B recoil charge states could be analyzed simultaneously on the focal plane of the spectrograph, observation of $^{11}\text{B}^{2+}$ recoils was not routinely made. -Enough observations were made, however, to insure that there was no rapidly changing behavior of the $^{11}\text{B}^{2+}$ charge-state abundance as a function of target thickness.

In Table BI the fractions of $^{11}\text{B}^{3+}$, $^{11}\text{B}^{4+}$ and $^{11}\text{B}^{5+}$ recoils at 11.1×10^8 cm/sec appear for 15 natural boron targets ranging in thickness from 0.03 to $54.20 \mu\text{g cm}^{-2}$. In computing the fractions the number of $^{11}\text{B}^{2+}$ recoils was taken as zero. In Table BII all four charge-state fractions of ^{11}B recoils at 11.1×10^8 cm/sec appear for the five natural boron targets for which the $^{11}\text{B}^{2+}$ recoil spectrum was observed. Comparison of Tables BI and BII demonstrates that the differences in the charge-state abundances by inclusion of the $^{11}\text{B}^{2+}$ abundance is well within statistical error. The charge-state fractions of Table BI are plotted against thickness in Figure B1.

From Figure B1 it can be seen that the $^{11}\text{B}^{3+}$ charge-state fraction decreases with decreasing target thickness while the $^{11}\text{B}^{5+}$ charge-state fraction increases with decreasing target thickness. The $^{11}\text{B}^{4+}$ charge-state fraction appears to be independent of target thickness within the error bars. This behavior is similar to that of the ^{12}C recoil charge-state fractions at 8.4×10^8 cm/sec from $^{12}\text{C}(\text{d},\text{d})^{12}\text{C}$ elastic scattering [Wolke, et al., Phys. Rev. Lett. 27, 1449 (1971), See Fig. C1] even though a different number of electrons

Table BI

Target Number	Thickness ($\mu\text{g}\cdot\text{cm}^{-2}$)	Charge-State Fractions		
		$Y^*(3)$ (%)	$Y^*(4)$ (%)	$Y^*(5)$ (%)
B004	54.20 $\pm$ 8.01	27.61 $\pm$ 0.49	54.49 $\pm$ 0.75	17.90 $\pm$ 0.38
B112	40.79 $\pm$ 6.03	28.74 $\pm$ 0.50	53.53 $\pm$ 0.75	17.73 $\pm$ 0.38
B101	30.08 $\pm$ 4.45	28.87 $\pm$ 0.50	53.36 $\pm$ 0.74	17.77 $\pm$ 0.37
B105	23.38 $\pm$ 3.46	27.76 $\pm$ 0.58	54.04 $\pm$ 0.90	18.20 $\pm$ 0.45
B424	12.63 $\pm$ 1.87	27.90 $\pm$ 0.59	53.84 $\pm$ 0.86	18.27 $\pm$ 0.43
B421	11.35 $\pm$ 1.68	26.57 $\pm$ 0.53	54.44 $\pm$ 0.69	18.99 $\pm$ 0.35
B420	9.70 $\pm$ 1.43	28.70 $\pm$ 0.58	52.78 $\pm$ 0.82	18.52 $\pm$ 0.43
B423 [†]	8.50 $\pm$ 1.26	27.13 $\pm$ 0.52	53.78 $\pm$ 0.75	19.08 $\pm$ 0.39
B223 [†]	8.08 $\pm$ 1.19	27.05 $\pm$ 0.61	53.92 $\pm$ 0.92	19.04 $\pm$ 0.47
B256 [†]	2.75 $\pm$ 0.41	25.48 $\pm$ 0.62	53.18 $\pm$ 0.81	21.34 $\pm$ 0.45
B455 [†]	1.31 $\pm$ 0.20	22.52 $\pm$ 0.75	54.40 $\pm$ 1.07	23.07 $\pm$ 0.62
B251 [†]	0.52 $\pm$ 0.08	21.98 $\pm$ 0.74	53.59 $\pm$ 1.00	24.43 $\pm$ 0.59
B452	0.33 $\pm$ 0.03	19.36 $\pm$ 1.17	55.26 $\pm$ 1.38	25.38 $\pm$ 0.81
B450	0.21 $\pm$ 0.02	19.95 $\pm$ 1.73	56.73 $\pm$ 2.02	23.32 $\pm$ 1.01
B458	0.03 $\pm$ 0.01	13.75 $\pm$ 5.73	59.48 $\pm$ 5.72	26.77 $\pm$ 3.13

[†]Without inclusion of $^{11}\text{B}^{2+}$ charge-state fraction - see Table BII.

Errors shown are statistical standard deviation.

Table BII

Target Number	Thickness ($\mu\text{g}\cdot\text{cm}^{-2}$)	Charge-State Fractions			
		$Y^*(2) (\%)$	$Y^*(3) (\%)$	$Y^*(4) (\%)$	$Y^*(5) (\%)$
B423	8.50 ± 1.26	1.71 ± 0.16	26.67 ± 0.51	52.86 ± 0.73	18.76 ± 0.38
B223	8.08 ± 1.19	1.07 ± 0.24	26.75 ± 0.60	53.34 ± 0.91	18.84 ± 0.47
B256	2.75 ± 0.41	1.81 ± 0.23	25.00 ± 0.61	52.21 ± 0.80	20.93 ± 0.44
B455	1.31 ± 0.20	2.58 ± 0.29	22.39 ± 0.74	54.09 ± 1.07	22.94 ± 0.62
B251	0.52 ± 0.08	$\leq 0.87^\dagger$	21.98 ± 0.74	53.59 ± 1.00	24.43 ± 0.59

† Only maximum possible value can be assigned as subtraction of normalized Ni blank resulted in negative counts.

Errors shown are statistical standard deviation.

Figure B1

is involved for the same charge state. (A $^{11}\text{B}^{5+}$ recoil is completely stripped of its electrons while a $^{12}\text{C}^{5+}$ recoil retains one electron. This is in contrast to the behavior of the ^{10}B recoil charge-state fractions at 11.1×10^8 cm/sec from the $^{12}\text{C}(d,\alpha)^{10}\text{B}$ reaction [NYO-2771-40 and COO-3427-1, See Fig. C2] where the thinnest target ($0.15 \mu\text{g}\cdot\text{cm}^{-2}$) showed a sudden decrease in the $^{10}\text{B}^{4+}$ charge-state abundance, resulting in a maximum value in the charge-state fraction versus target thickness data, and showed a sudden increase in the $^{10}\text{B}^{3+}$ charge-state abundance resulting in a minimum value in the charge-state fraction versus target thickness data. It was intended to observe ^{11}B charge-state distributions from targets less than $0.20 \mu\text{g}\cdot\text{cm}^{-2}$ thick in order to determine whether the sudden change in abundances of the ^{10}B charge-state fractions was unique. As shown in Table B1, charge-state distributions from two targets in the thickness range $\leq 20 \mu\text{g}\cdot\text{cm}^{-2}$ fail to exhibit any sudden changes ($0.03 \mu\text{g}\cdot\text{cm}^{-2}$ is less than one atomic layer thick on the average) in the ^{11}B charge-state fraction versus thickness data.

In terms of the model published earlier, this supports the premise that either the sudden change in nuclear charge which occurs for the (d,α) reaction (coulomb shakeoff) or coulombic interactions with the electron cloud by the outgoing particle or some combination of the two is primarily responsible for the sudden change in charge-state abundances observed for ^{10}B recoils from the $^{12}\text{C}(d,\alpha)^{10}\text{B}$ reaction, as both the coulombic interaction of the incoming projectile with the target atom's electron cloud and the sudden recoil of the residual nucleus (recoil shakeoff) should be

similar for ^{10}B recoils from the $^{12}\text{C}(\text{d},\alpha)^{10}\text{B}$ reaction and ^{11}B recoils from $^{11}\text{B}(\text{d},\text{d})^{11}\text{B}$ elastic scattering at 11.1×10^8 cm/sec.

2. The $^{12}\text{C}(\text{d},\alpha)^{10}\text{B}$ Reaction

Equilibrium and pre-equilibrium charge state distributions (CSD's) of recoils from the $^{12}\text{C}(\text{d},\alpha)^{10}\text{B}$ reaction in graphite targets were reported upon previously. Efforts to obtain additional ^{10}B CSD's in the $< 1 \mu\text{g}/\text{cm}^2$ region continue to be hampered by carbon contamination from pump oil residues in the spectrograph scattering chamber.

Measurement of the equilibrium CSD of ^{10}B recoils from the $^{12}\text{C}(\text{d},\alpha)^{10}\text{B}$ reaction using diamond targets, however, has been undertaken during the past year. If the ^{10}B recoil velocity is identical to the velocity of the ^{10}B recoils from graphite targets of comparable thickness, a comparison of the equilibrium CSD from these two targets might show a dependence on the atomic structure of the target medium. Such a "medium effect" between diamond and graphite might result from differences in Auger electron emission from excited ions or from differences in the electron capture and loss interactions in the two media. Such differences might arise from differences in electron density and crystal structure between the two forms of carbon.

Targets of powdered diamond were prepared (See Section F2) on 0.1 mil Ni foil backings and irradiated with 11.40 MeV deuterons produced in the Van de Graaff accelerator. Graphite targets of comparable thickness were irradiated in the same manner. Boron-10 recoils were observed at 11.1×10^8 cm/sec at 20° to the beam from the $^{12}\text{C}(\text{d},\alpha)^{10}\text{B}$ reaction in both targets and were analyzed in the

Enge Split-Pole Spectrograph. The experimental arrangement for these measurements has been reported previously.

Equilibrium CSD's of ^{11}B recoils measured in natural boron targets had previously shown no statistically significant difference from the equilibrium CSD's of ^{10}B recoils in graphite targets when the recoil velocity is 11.1×10^8 cm/sec. In order to detect the existence of any "medium effect" in the present experiment, therefore, counting statistics of less than 0.5% must be obtained from both the carbon and the diamond targets.

Irreproducibilities in experimental parameters may account for a small difference observed between the ^{10}B CSD's from diamond and graphite targets bombarded some weeks apart. It is necessary that the data be accumulated from both targets over a period of days. Since the total number of recoils produced per unit target thickness and per microcoulomb of beam for graphite targets is known from our previous work, the use of graphite targets of known thickness provides an internal check on the consistency of the CSD data.

Preliminary experiments have shown the ^{10}B recoil baseline peak widths (3+, 4+, and 5+ peaks) on the focal plane of the Enge Split-Pole Spectrograph from diamond targets to be 1-2 cm larger than those from graphite targets of comparable thickness. This may indicate that the actual thicknesses of matter on the diamond targets is significantly greater than the graphite target thicknesses, resulting in an increased total energy loss of the ^{10}B recoils in the diamond targets. Since the blanks prepared show no visible contamination from the water used to suspend the diamond dust (see Section F2), indications are that

a substantial contaminant may be present in the diamond dust. Efforts to isolate and correct the problem are in progress.

3. The $^{11}\text{B}(\text{d},\alpha)^9\text{Be}$ Reaction

Experiments on the pre-equilibrium CSD's of recoils from the $^{11}\text{B}(\text{d},\text{d})^{11}\text{B}$ reaction at 11.1×10^8 cm/sec were designed to examine the contribution of coulomb shakeoff (and/or the contribution of coulombic interaction with the electron cloud by the outgoing particle) to the "instantaneous" or primary charge distribution. If the recoil velocity were to be changed significantly, measurement of the pre-equilibrium charge distribution as a function of target thickness might be expected to shed further light on the contribution of coulomb shakeoff to primary charge distributions.

The $^{11}\text{B}(\text{d},\alpha)^9\text{Be}$ reaction at a deuteron bombarding energy of 16.90 MeV, which yields a ^9Be recoil of 15.20 MeV (velocity 18.0×10^8 cm/sec) at a scattering angle of 15° , was chosen for this purpose. Although the reaction cross section is an order of magnitude lower than the $^{11}\text{B}(\text{d},\text{d})^{11}\text{B}$ reaction at a deuteron energy of 14.5 MeV, the kinematic broadening contribution to the baseline width of the ^9Be recoil peak at the focal plane of the spectrograph is approximately equal to that of the ^{11}B peak. Consequently, the horizontal angle of acceptance could be opened to $> 1^\circ$, producing a compensating increase in the recoil counting rate.

Due to the high ^9Be recoil velocity, only two charge states are expected to appear in the CSD. The absence of a third charge state is not only experimentally attractive but may also result in a more easily-handled calculational system in the calculation of primary charge state distributions.

No target development work was necessary for this experiment. Techniques used in the fabrication and calibration of boron targets have been routinely used during the past year.

Results from preliminary measurements of the equilibrium CSD of ^9Be recoils from the $^{11}\text{B}(d,\alpha)^9\text{Be}$ reaction show that the two charge states $4+$ and $3+$ are present. Preliminary measurements indicate that the $2+$ state abundance is less than 1%. The number of recoils per unit target thickness per microcoulomb of beam collected shows the ^9Be counting rate to be comparable to that obtained for ^{10}B recoils in the $^{12}\text{C}(d,\alpha)^{10}\text{B}$ reaction. Measurements on the $^{11}\text{B}(d,\alpha)^9\text{Be}$ reaction are continuing.

C. Calculation of Charge-Changing Cross-Sections

The interactions of an ion beam with a solid or gaseous target can be described by the system of linear coupled differential equations

$$dY(q)/dx = \Sigma[\sigma(q',q)Y(q') - \sigma(q,q')Y(q)], \quad (1)$$

where $Y(q)$ is the pre-equilibrium charge-state fraction of charge q ,

$\sigma(q,q')$ is the cross-section for electron capture or loss by

the ground state ion of charge q . (Note that: $q > q'$

for electron capture; $q < q'$ for electron loss),

x is the target thickness.

Here $Y(q)$ is a function of Z the atomic number of the impinging ion, v its velocity, x , and Z_T the atomic number of the target material.

If equilibration has been achieved, the system (1) becomes

$$\Sigma[\sigma(q',q) F(q') - \sigma(q,q') F(q)] = 0 \quad (2)$$

where $F(q)$ refers to the equilibrium charge-state fraction of charge q and is a function of Z , v and Z_T .

For recoiling ions from nuclear processes, the system of differential equations (1) cannot be applied without modification. The number of recoils of charge q leaving some target thickness element dx can be expressed as $dN(q)$ and is made up of three terms: the number of recoils of charge q formed in dx by the nuclear process plus the number of recoils of charge q formed in dx from recoils of charge q' entering from previous thickness elements, minus the number of recoils of charge q which are transformed within dx into recoils of charge q' :

$$dN(q) = \sigma_q N'_p dx + \sum_{q'} [\sigma(q', q) N'(q') - \sigma(q, q') N'(q)] dx, \quad (3)$$

where σ_q is the cross-section for the nuclear process $V(a, b)W^q$,

N'_p is the number of beam particles entering the element dx ,

$N'(q)$ is the number of recoils of charge q entering the element dx ,

$N(q)$ is the number of recoils of charge q leaving the element dx .

Since the target is only a few $\mu\text{g cm}^{-2}$ thick, $N'_p = N_p$, and since the element dx is infinitesimal, $N'(q) \approx N(q)$. Then

$$dN(q)/dx = \sigma_q N_p + \sum_{q'} [\sigma(q', q) N(q') - \sigma(q, q') N(q)]. \quad (4)$$

As $\sum_q \sigma_q = \sigma$,

$$\sum_q N(q) = \sigma N_p x. \quad (5)$$

Taking the derivative of (5) yields

$$d[\sum_q N(q)] = \sigma N_p dx. \quad (6)$$

The detected recoil charge-state fractions $Y^*(q)$ and $F^*(q)$ differ from those described in Section 1.1 since in the present case they are a sum of charge-state fractions produced at different depths in the target. In terms of the observable,

$$Y^*(q) = N(q)/\sum_q N(q). \quad (7)$$

Taking the derivative of (7) yields

$$dY^*(q) = [\sum_q N(q)]^{-1} dN(q) - N(q) [\sum_q N(q)]^{-2} d[\sum_q N(q)]. \quad (8)$$

Substituting (5), (6) and (7) into (8) and rearranging gives

$$dN(q)/dx = [\sum_q N(q)] [dY^*(q)/dx + Y^*(q)/x]. \quad (9)$$

Equating (9) and (4), and using the definition (7) yields

$$dY^*(q)/dx + Y^*(q)/x = \frac{1}{x} [\sigma_q/\sigma] + \sum_{q'} [\sigma(q',q)Y^*(q') - \sigma(q,q')Y^*(q)]. \quad (10)$$

Since σ_q/σ is $Y^0(q)$ the primary charge-state fraction,

$$dY^*(q)/dx + Y^*(q)/x = Y^0(q)/x + \sum_{q'} [\sigma(q',q)Y^*(q') - \sigma(q,q')Y^*(q)]. \quad (11)$$

Now a least squares fit to the experimental data of a numerical integration of the system (11) should yield the electron capture-loss cross-sections. As discussed by Betz [Rev. Mod. Phys. 44, 465 (1972)], these cross-sections should be more properly designated $\sigma^*(q,q')$ as they will reflect residual ion excitation of the recoils in the solid and an undetermined amount of Auger electron emission by the recoils upon leaving the target medium:

$$\sigma^*(q,q') = \sum_i a_i \sigma_i(q,q') + \sigma_A, \quad (12)$$

where $i = 0$ refers to the ground state, $i = 1$ refers to the first excited state, etc.; a_i is the fraction of ions of charge q in state i ; and σ_A is the undetermined Auger emission addition.

Electron capture-loss cross-sections were calculated for three reactions: the data of Section B1, the data of Wolke, et al. [Phys. Rev. Lett. 27, 1449 (1971)] and the data of COO-3427-1. The computer program RAVEX was used for these calculations. RAVEX accepts a "guess set" of cross-sections and numerically integrates the system (11) for that set using a Runge-Kutta-Gill numerical integration. As the numerical integration requires a value for the primary or "instantaneous" charge-state distribution, the charge-state distribution from the thinnest target in each case was used. Two main logic

patterns are applied by RAVEX in arriving at a weighted least-squares sum of the differences between the calculated curves and the experimental pre-equilibrium data. The first logic pattern varies the value of each cross-section by some fraction of that value, independently of the values of the other cross-sections, and examines the effect of this variation on the differences-squared sum. As the differences-squared sum decreases, the variance in a cross-section which will allow a further decrease becomes smaller. The second logic pattern is independent of the first and uses time of execution as a guide. Periodically, current values of the cross-sections are compared with their values at the beginning of the time period. A prediction then is made as to what their values would be after another period based on the rate of change of each cross-section, and the current values are correspondingly adjusted. When only changes of less than 0.1% in the value of each cross-section can be recorded, execution ceases. Any cross-section value which becomes less than $10^{-6} \text{ cm}^2/\mu\text{g}$ is reset to zero to help minimize roundoff error in the calculations and decrease the execution time. The value of such a cross-section is thus not determined and can only be said to be less than $10^{-6} \text{ cm}^2/\mu\text{g}$ (see below). The value of RAVEX lies in reasonably fast convergence of the cross-section values when the "guess set" of values is in error by a factor of 10^5 .

The cross-sections are presented in Table CI in units of $\text{cm}^2/\mu\text{g}$ and in Table CII in units of $10^{-16} \text{ cm}^2/\text{atom}$. Figures B1, C1 and C2 show the calculated curves fitted to the experimental data for ^{11}B recoils at $11.1 \times 10^8 \text{ cm/sec}$ in boron, ^{10}B recoils at $11.1 \times 10^8 \text{ cm/sec}$ in graphite and ^{12}C recoils at $8.4 \times 10^8 \text{ cm/sec}$ in graphite

Table CI

Reaction	Recoil Velocity ($\times 10^8$ cm/sec)	Target Medium	Electron Capture-Loss Cross-Sections ($\text{cm}^2/\mu\text{g}$)					
			$\sigma^*(3,4)$	$\sigma^*(3,5)$	$\sigma^*(4,3)$	$\sigma^*(4,5)$	$\sigma^*(5,3)$	$\sigma^*(5,4)$
$^{11}\text{B}(d,d)^{11}\text{B}$	11.1	boron	4.1	$\sim 0.01^\dagger$	2.0	0.6	$< 10^{-6}^\ddagger$	1.7
$^{12}\text{C}(d,\alpha)^{10}\text{B}$	11.1	graphite	10.9	1.3	5.2	$< 10^{-6}^\ddagger$	$< 10^{-6}^\ddagger$	1.4
$^{12}\text{C}(d,d)^{12}\text{C}$	8.4	graphite	1.6	$< 10^{-6}^\ddagger$	0.7	0.3	$< 10^{-6}^\ddagger$	1.1

† Error may be as great as a factor of 10 (see text).

‡ Not determined (see text).

Table CII

Reaction	Recoil Velocity ($\times 10^8$ cm/sec)	Target Medium	Electron Capture-Loss Cross-Sections ($\times 10^{-16}$ cm ² /atom)					
			$\sigma^*(3,4)$	$\sigma^*(3,5)$	$\sigma^*(4,3)$	$\sigma^*(4,5)$	$\sigma^*(5,3)$	$\sigma^*(5,4)$
$^{11}\text{B}(d,d)^{11}\text{B}$	11.1	boron	0.74	$\sim 2 \times 10^{-3}^\dagger$	0.36	0.11	$< 2 \times 10^{-7}^\ddagger$	0.31
$^{12}\text{C}(d,\alpha)^{10}\text{B}$	11.1	graphite	2.17	0.26	1.04	$< 2 \times 10^{-7}^\ddagger$	$< 2 \times 10^{-7}^\ddagger$	0.28
$^{12}\text{C}(d,d)^{12}\text{C}$	8.4	graphite	0.32	$< 2 \times 10^{-7}^\ddagger$	0.14	0.06	$< 2 \times 10^{-7}^\ddagger$	0.22

[†]Error may be as great as a factor of 10 (see text).

[‡]Not determined (see text).

Figure C1

Figure C2

respectively. These curves were generated by the computer program ODIN. ODIN accepts a least-square-determined set of electron capture-loss cross-sections and numerically integrates and plots the system (11) using a Runge-Kutta-Gill numerical integration and a Calcomp plotter. The data points are also plotted by ODIN for comparison with the calculated curves.

The values quoted in Tables CI and CII are considered to be preliminary for the experimental data shown, since the final logic checks for RAVEX are not yet complete. In addition:

- (a) Values given as "less than some number" could not be determined from the experimental data due to extremely weak interaction with the system (11).
- (b) Values given as " $\sim$ " may be inaccurate by as much as a factor of 100 due to weak interaction with the system (11).

Errors of the order of $\pm 50\%$ of the given cross-section values may be possible if the primary charge-state distribution is not fairly approximated by the charge-state data from the thinnest target in each case.

There is similarity in the capture cross-section $\sigma^*(5,4)$ for all three recoil systems shown in Tables C1 and C2 even though a "second" electron is being captured by the $^{12}\text{C}^{5+}$ recoil as opposed to a "first" electron being captured by the B^{5+} recoils. The largest cross-section for all three recoil systems is the loss cross section $\sigma^*(3,4)$, and there is evidence that double capture or loss plays little part in determining the equilibrium charge-state fractions with the exception of the double loss cross-section $\sigma(3,5)$ for ^{10}B .

recoils. This value may be in error, as may be all the ^{10}B cross-sections, as evidenced by the difficult fit of the calculated curves to the experimental data points. As the ^{10}B recoil charge-state data are similar to those for the ^{11}B and ^{12}C recoils except those from the thinnest target, it would be expected that cross-sections similar to those quoted for the ^{11}B and ^{12}C recoils would result from the calculations if the data from the thinnest target were omitted. However, as no experimental reason exists for doubting the validity of the data from that target (C00-3427-1) such a calculation has not been done. It is possible that the capture-loss cross-sections cannot adequately reflect the behavior of the ^{10}B charge-state distribution from the $0.15 \mu\text{g cm}^{-2}$ target when the system (11) is numerically integrated.

D. Tritium Enrichment in Water

Proposed for 1972 was a project to develop a novel method for the isotopic enrichment of tritium in water. The method, based on the low-temperature electrolysis of water dissolved in liquid sulfur dioxide, was originally conceived of as producing an automatic multistage subjection of the same water molecules to electrolytic discharge, an effect which could be expected to produce large enrichment factors. Shortly after beginning this work it was realized that our reasoning had been faulty and that the system was capable at most of producing a single stage electrolysis of the water, albeit at a much lower temperature than could be used in a completely aqueous system and hence with the possibility of achieving a higher separation factor inasmuch as low temperatures enhance the isotopic differences.

Solutions of tritiated water of known specific activity plus bromine in liquid sulfur dioxide were electrolyzed at -75°C between platinized platinum electrodes in a specially-constructed electrolytic cell. Bromine is necessary in the system to produce sufficient conductivity by reaction with the water. The water remaining in the cell after the electrolysis and after the SO_2 and Br_2 were removed was purified and counted to determine its specific activity and hence the degree of isotopic enrichment. Electrolyses were carried out under a variety of conditions of concentration, voltage and current. All of the results showed little if any isotopic enrichment, however, most likely because of the interference of chemical reactions within the system, such as the oxidation of SO_2 to SO_4^- by bromine.

E. Computation

During the past year our use of computers has increased substantially as new programs for calculations and data analysis have been developed and employed.

The University of Pittsburgh ceased operation of its IBM-7090 and IBM 360/50 system on schedule. The conversion of our programs, formerly compatible with the IBM computers and PDP-15 computer, to the PDP-10 has been completed. The magnetic tape reading and printing package, the matrix inversion program, the γ - γ angular correlation matrix stripping and printing package, and the relativistic kinematics programs are currently operational on the PDP-10. In addition, MONTY2, a non-linear weighted polynomial least-squares program was developed and made operational on the PDP-10 computer.

A number of programs are being developed in order to facilitate data handling and analysis:

- 1) A plotting package is being developed which is designed to plot any specified function and/or any specified array of x, y points.
- 2) A program, RAVEX, has been designed to determine a complete set of $\sigma(q, q')$'s from pre-equilibrium charge-state data from nuclear reactions. The program uses a least-squares fit of a Runge-Kutta-Gill numerical integration to the data points as discussed in C above.
- 3) A package, ODIN, has been programmed which will plot experimental charge state data points and their associated errors and will plot a least squares fitted curve to the $\sigma(q, q')$'s as discussed in C above.

F. Targetry

1. Boron Targets

In the past year, all of the boron targets required for the work in Sections B1 and B3 above have been made by the method described last year for the production of thick boron targets.

The optical characteristics of boron films deposited on 0.0001 inch nickel, as reported last year, continue to be of incidental interest. The intense, bright colors of these films appear to be directly related to their thicknesses. With increasing thickness the colors of the films follow the sequence: yellow, blue, red, gray, green, violet. This correlation with thickness can be seen not only by comparison of films of known thicknesses, but also by observation of the film buildup during the evaporation process. As the boron is deposited on the foil, the colors change according to the above sequence. Apparently, something more than simple light interference is involved. The possibility of using the color-thickness correlation as a thickness measurement is being investigated.

For the first part of the year, our target-making activities were severely hindered by equipment malfunctions.

- a) The Cahn Gram Electrobalance, which had been used as the primary calibration instrument for weighed target thickness determinations, became increasingly erratic and irreproducible. Attempts by Cahn to repair the balance were not successful. Since accurate weighed thickness determinations are crucial to

our work, a model 4125 Sartorius Electronic Microbalance was acquired to replace the Cahn.

- b) The sensor head of the quartz crystal microbalance system became quite unreliable this year and was replaced with a model 103-926 Remote Crystal Holder from Sloan Technology Corporation. The design of this sensor head is somewhat different from the design of the one used previously, but it is completely compatible with the quartz crystal microbalance electronics now in use.

An independent method of determining target thicknesses for boron targets above $0.5 \mu\text{g}/\text{cm}^2$ has been developed. Deuterons at 10.00 MeV are scattered elastically from the boron targets at 50° and detected on the focal plane of the Enge Split-Pole Spectrograph by a 12 mm-diameter, partially depleted, silicon surface barrier detector operated at an 800 μ depletion depth. The number of elastically scattered deuterons was calibrated against six weighed boron films as shown in Table FI. This calibration value of 90 ± 133 elastically scattered deuterons per $\mu\text{g}/\text{cm}^2$ per 100 μcoul of incident deuterons was used to establish the target thicknesses of all targets thicker than $0.5 \mu\text{g}/\text{cm}^2$ as shown in Table FII. Background from the nickel backing prevents the determination by deuteron elastic scattering of boron thicknesses less than $0.5 \mu\text{g}/\text{cm}^2$.

Target thicknesses below $0.5 \mu\text{g}/\text{cm}^2$ are therefore determined from the number of ^{11}B recoils produced per $\mu\text{g}/\text{cm}^2$ per 5000 microcoulombs of incident deuterons (2024 ± 166).

Table FII - Calibration of Elastically Scattered Deuterons Against Weighed Boron Targets

Target Number	Thickness Determined by Weighing ($\mu\text{g}\cdot\text{cm}^{-2}$)	Number of Elastic Deuterons per $\mu\text{g}\cdot\text{cm}^{-2}$ per 100 μcoul of incident deuterons	Number of ^{11}B recoils per $\mu\text{g}\cdot\text{cm}^{-2}$ per 5000 μcoul of incident deuterons
B420	$11.46 \pm 0.48^*$	762	2202
B421	11.56 ± 0.49	884	2090
B422	13.12 ± 0.50	995	
B423	10.59 ± 0.28	722	1811
B424	11.37 ± 0.44	1000	1991
B425	13.80 ± 0.46	1036	
Average		$900 \pm 133^*$	$2024 \pm 166^*$

*Errors are one standard deviation

Table FIII - Boron Targets

<u>Target Number</u>	<u>Thickness*</u> <u>($\mu\text{g}\cdot\text{cm}^{-2}$)</u>
B004	54.20 ± 8.01
B112	40.79 ± 6.03
B101	30.08 ± 4.45
B105	23.38 ± 3.46
B424	12.63 ± 1.87
B421	11.35 ± 1.68
B420	9.70 ± 1.43
B423	8.50 ± 1.26
B223	8.08 ± 1.19
B256	2.75 ± 0.41
B455	1.31 ± 0.20
B251	0.52 ± 0.08
B452	$0.33 \pm 0.03^\dagger$
B450	$0.21 \pm 0.02^\dagger$
B458	$0.03 \pm 0.01^\dagger$

*Based on 900 ± 133 elastic deuterons per $\mu\text{g}\cdot\text{cm}^{-2}$
per 100 μcoul of incident deuterons

† Based on 2024 ± 166 ^{11}B recoils per $\mu\text{g}\cdot\text{cm}^{-2}$ per
5000 μcoul of incident deuterons

2. Diamond Targets

Twenty carats of 0-0.25 micron diamond dust were obtained from Ace Abrasive Lab for use in target making. Two methods were employed in attempts to produce the thin, uniform diamond targets needed for the work described in Section B2, above. The first method consisted of suspending the diamond dust in deionized, doubly distilled water to which a drop of a 1% Aerosol OT solution had been added to reduce surface tension. The larger diamond particles were permitted to settle out. The remaining suspension was transferred to 0.001 inch nickel foils in small quantities with a Pasteur pipet and evaporated under a heat lamp. The targets produced in this way, however, were non-uniform.

The second method involved spraying a fine mist of aqueous diamond dust suspension onto the nickel foil. The suspension was first centrifuged for a few minutes to remove the larger particles. The resulting supernate consisted of very fine particles in a suspension which lasted for several days. This suspension was sprayed onto the nickel foil by means of a model 642 DeVilbiss Nebulizer, utilizing a stream of pure dry nitrogen. During the nebulizing operation, the foil was rotated and heated to insure uniform deposition of the diamond. The resulting targets, when examined under a 400X microscope, appeared to be quite uniform. Although accurate thickness determinations have not yet been made, it is felt that uniform targets can be made in thicknesses varying from equilibrium down into the pre-equilibrium range.

3. Wherrett Evaporator System

The current status of the Wherrett Evaporator System remains as reported last year. Funds were not available for further construction this year, and work on the evaporator has therefore been suspended.

III. PERSONNEL

Principal Investigator

Robert L. Wolke, Professor of Chemistry, B.S. Chem. Polytechnic Institute of Brooklyn 1949, Ph.D. Cornell University 1953. (Approximately 40% to date; approximately 20% January-March 1973.)

Graduate Research Assistants

Ted W. Debiak, B.S. Pennsylvania State University 1966. (Through August 1972. NSF Center-of-Excellence Fellow, September 1972 to present.)

Marghoob Ahmad Khan, B.S. Peshawer University 1961. (Half-time.)

Edward V. Mason, Jr., B.S. University of Richmond 1967. (Through August 1972. Teaching Assistant, September 1972 to present.)

John D. Yesso, B.S. University of Pittsburgh 1971. (Through August 1972. Teaching Assistant, September 1972 to present.)

Undergraduate Assistant

William J. Jordan

Secretary

Lillian M. Abernethy